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A New Solid State Tritium Surface Monitor

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Abstract

Traditionally the amount of tritium on a surface is determined by swiping the surface with a material such as filter paper and counting the removed tritium by scintillation. While effective, this method can be time consuming, can alter the surface and only measures removable tritium. For a given application each of these considerations may or may not be a disadvantage. A solid state monitor, on the other hand, has the potential to provide rapid analysis, not alter the surface and measure all tritium on a surface. This allure has promoted open wall ion chamber and PIN diode-based tritium surface monitor development, and these techniques have enjoyed certain success. Recently the first tests were performed with an avalanche photodiode (APD) for surface tritium measurement. The tritium surface concentration is determined by placing the APD within a few millimeters of the surface of interest. Beta decay from the surface tritium impacts the APD resulting in amplified current through the diode. Analysis of this signal with a multi-channel analyzer enables counting of beta decay events and determination of the beta energy spectrum. While quite similar in concept to PIN diode based measurements, side-by-side testing showed that the APD provided substantially better counting efficiency. Considerations included count rate, background, sensitivity, stability and effect of ambient light. An important factor in the U.S. for a tritium surface monitor is the ability to measure concentrations down to the "free release" limit, i.e., the concentration below which items can be removed from radiological control areas. The two limits being used are 10,000 disintegrations per min (dpm)/100 cm^2 and 1,000 dpm/100 cm^2. Present tests show that the APD is capable of measuring down to 1,000 dpm/100 cm^2 in reasonable count times. Data from this promising technique will be presented in this paper.

Introduction

A key component of radiological control in tritium facilities is measurement of tritium surface concentrations. This is needed to prevent the spread of contamination, determine appropriate personnel protective equipment and to assess the effectiveness of decontamination measures. Its use is particularly important at radiological boundaries to determine whether or not items can be cleared (released for use) outside of the facility. Currently this function is accomplished by swiping the item and measuring the removed tritium with scintillation counting. This method can be time consuming, especially when centralized facilities are used for counting. And this method produces waste.

To clear material out of a tritium facility, the material's tritium surface concentration must be measured and shown to be below the free release limit. For many years the free release limit for DOE tritium facilities was 1000 disintegrations per minute per 100 cm^2 (dpm/100 cm^2). Not long ago, DOE increased the free release limit to

10,000 dpm/100 cm^2. However, to maintain an added level of safety, some DOE facilities maintain local free release limits at 1000. Thus, there two free release limits currently in effect. A tritium surface characterization technique will be most useful for use in US DOE facilities if it is capable of accurate measurements at both levels.

For this purpose, solid state tritium detectors have potential advantages over the current swipe/scintillation method. Solid state instruments can be direct-reading, rapid, simple-to-use and waste-free. In earlier work Wampler and Doyle [1] showed that a PIN diode-based system, generally used for x-ray measurements, could be used for tritium measurements by removing the usual beryllium window over the diode. However, this method has not yet been shown to be sensitive to free release concentrations. Avalanche Photodiodes (APD's), on the other hand, were believed to have the potential to accurately measure tritium surface concentrations at the free release limit, and this motivated the present work. The first work on APD's was over thirty years ago by Johnston, et al. [2]. And they have been considered for use as various types of tritium detectors by McGann, et al. [3], Surette [4], and Shah, et al. [5]. These references give detailed physical descriptions of APD's so such information will not be repeated here.

The present work was dedicated to evaluating the suitability of an APD-based system as a practical technique for measuring tritium surface concentrations at or near the free release limits. This work also has broader implications such as scientific studies which need non-invasive methods for measuring tritium surface concentrations.

Experimental Setup

A 77 mm^2, square-shaped avalanche photodiode (APD) was manufactured by RMD, Inc. for testing. This device is an experimental prototype so no model or part number is associated with it. RMD, Inc. also manufactured a prototype electronics package for the APD. This package supplied an adjustable voltage to the APD and collected the pulsed signal from the APD. The package included a simple local readout of integrated counts. The APD signal from the electronics package was passed (gain-adjusted) to a separate multi-channel analyzer (Amptek MCA800A). The MCA integrated counts from the APD into 1024 channels. The electronics package gain was set so that the entire tritium beta spectrum (0 to ~18.6 keV) was captured by these channels. Information in the MCA was displayed and stored on a laptop computer running via the software package Pmca (version 3.0) supplied by Amptek.

To supply a known beta emission rate to the APD, three commercially-availably tritium surface standards were used. The standards consisted of 100 cm^2 of porous aluminum with each standard containing a different quantity of tritium deposited in the porosity. The actually beta emission rate from the surface was characterized and certified by the manufacturer using NIST-traceable techniques. The three standards were selected to be nominally comparable to 100,000, 10,000 and 1,000 disintegrations per minute (dpm) per 100 cm^2. The latter two values correspond to "free release" limits for US Department of Energy (DOE) facilities. These values have considerable significance since material

found to be below the free release limits can be cleared for release outside DOE radiological facilities. The actual characteristics of the three plates used for this test are summarized in Table 1.

Table 1: Tritium Surface Standards Characteristics

Nominal Specific Activity (dpm/100 cm^2)	Specific Activity on Date of Test (dpm/100 cm^2)
100,000	85,440
10,000	11,700
1,000	1,000

While some solid-state detector require operation at low temperature, all of the tests reported here were successfully performed at room temperature.

Ambient light can result in a flood of signal from an APD, and indeed, the prototype APD was affected when exposed, for instance, directly to fluorescent room light. However, this was quite easily controlled. Thin, translucent plastic with an opening slightly larger than the APD was placed between the APD housing and the surface standard. Without further measures (e.g., shrouding or turning room light off), this was effective for light exclusion. The plastic also served to ensure that the APD and its housing did not become surface contaminated.

The plastic further served to ensure that the APD was always placed at a reproducible distance from the surface standard. Tritium beta particles have a quite limited range in air (1.82 mm for 5.68 keV beta in 1 atm air), thus it is important to place the APD as closely has possible to the beta source. In these test the distance was _____. While measures can be taken to decrease beta attenuation (e.g., remove air or use a less attenuating sweep gas) no measure of this kind was taken. Rather the tests were run at local atmospheric pressure air which in this case is 0.776 atm (78.6 kPa).

Need experimental arrangement figure here.

Figure 1 Experimental arrangement

Results

An example of the type of data collected from the APD is shown on figure 1. This is a counts vs. channel (energy) spectrum for the 85,440 dpm/100 cm^2 tritium standard plate. The counts were integrated over 69.8 hours. Extended counting was performed to collect significant counts in every channel to clearly show the shape of the collected spectrum. Four significant regions are visible in the spectrum. At the far left (channels 0-100) there are no counts shown. In reality there are a very large number of background counts in this region, but a threshold was set to ignore these counts thereby avoiding overloading the counting electronics. Moving to the right, say channels 100-200, the tail of the large background peak is visible. This peak is not indicative of the tritium concentration on the

plate. Rather, the spectrum associated with tritium is visible from approximately channel 300 to 900. This region has the general features of the known tritium spectrum shown in figure 3 [6]. Both the APD spectrum and the actual spectrum start with no signal at higher channel numbers (energy) and increase as the channel number decreases. However, the APD spectrum appears to peak at higher channel number (energy) than the actual spectrum. Causes for this might include air attenuation of the beta energy and non-linear response of the APD. The last region on figure 2 is at the highest channel numbers, say above 900, where there is no signal above background. An energy standard was not used to precisely convert APD channel number to energy, but it is noted that the APD signal is negligible at about channel number 900 and the actual tritium spectrum is negligible at ~18, so these values are taken to be roughly comparable. The APD system gain was set so that the APD signal became negligible at this high channel number, thus spreading the tritium spectrum across the entire range of channel numbers.

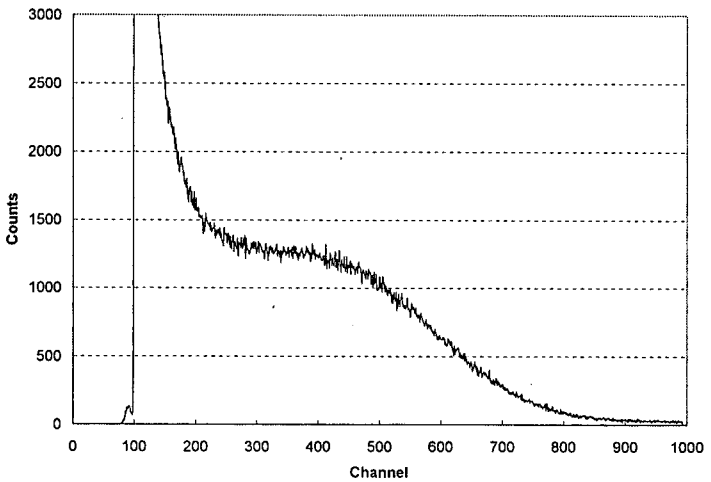

Figure 2 Counts vs. channel (energy) spectrum collected with prototype APD for 85,440 dpm/100 cm^2 plate over 69.8 hours

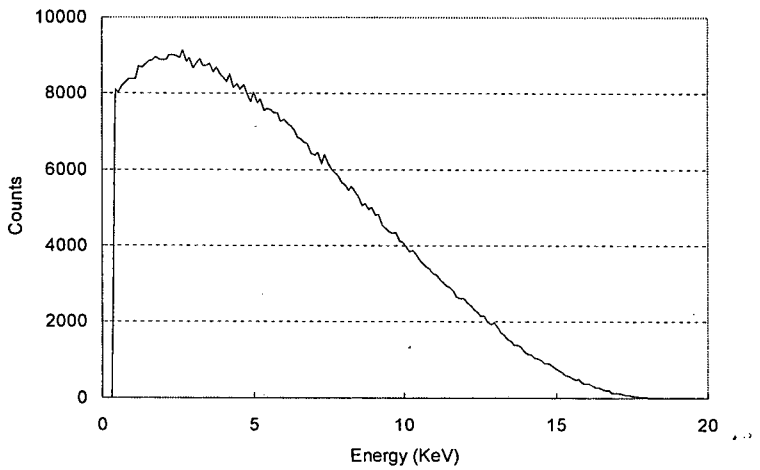

Figure 3 Actual tritium beta spectrum as given in Souers [6]

Extended counting was performed for all three tritium standard plates. Total counting time was used express the spectrum as a counting rate and the results are shown on figure 4. All three spectra show similar features.

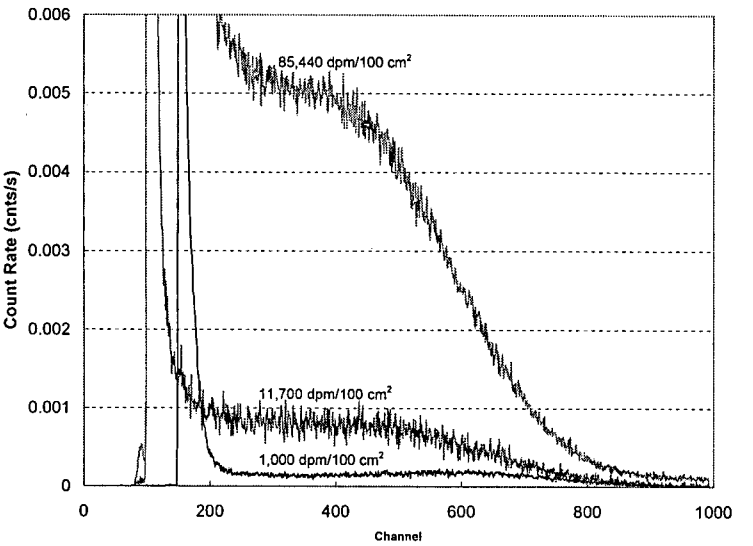

Figure 4 Spectrum for all three tritium standard plates expresses as counting rate

Proper quantification of surface tritium requires measurement of an APD system background. For this a sample of un-tritiated aluminum was read as if it were one of the tritium standard plates. The resulting background is shown on figure 5. As shown, the background counting rate is low. It is also evident that the background is not uniform across the region of interest; rather, it peaks at about channel number 700.

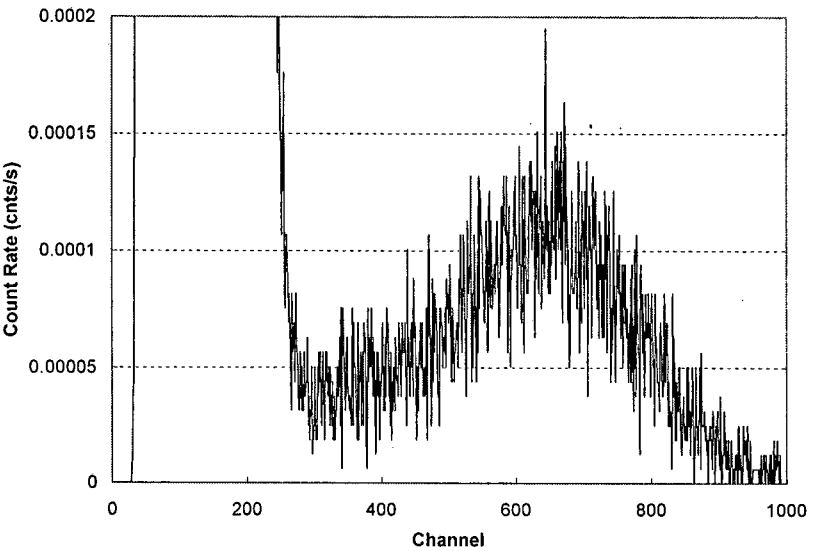

Figure 5 Background spectrum for aluminum plate

While the spectrum of betas is of interest, it is usually the total counts over all channels associated with tritium that would be used to quantify the surface tritium. Inspection of the tritium and background spectra indicates that the range 300-1000 (channel number)

appears to be reasonable for quantifying surface tritium. Thus, for the three surface standards and for the background, the counts in this range were summed and divided by the collection time. For the background, the result was 0.041 cnts/s. The results for the three standards are summarized in Table 2. As shown, the integrated count rates for the two highest surface standards are much larger than the background. For the lowest standard (1000 dpm/100 cm^2), the integrated count rate was 0.080 cnts/s—about twice the background. This should be sufficient signal over background for reliable measurements. However, measurements much below this would not be considered reliable. Nonetheless, it is important to note that the APD system has sufficient sensitivity to quantify tritium for both US free release levels.

Table 2 Summary of integrated count rate for three surface standards

Surface Standard (dpm/100 cm^2)	Count Time (hours)	Integrated Count Rate, CN 300-1000, (cnt/s)	Integrated Count Rate - Backround, CN 300-1000 (cnt/s)	Detection Efficiency (%)
85,440	69.8	1.577	1.536	14.0
11,700	19.4	0.291	0.250	16.7
1,000	96.4	0.080	0.039	29.3

Calculation of tritium surface concentration should be based on integrated count rate minus background count rate, so this quantity is also given in Table 2. These values were plotted versus the actual, total beta emission rate under the 77 mm^2 APD, and the result is shown as figure 6. The three experimental points fall nicely along a straight line which passes through the origin and has a slope of 0.141.

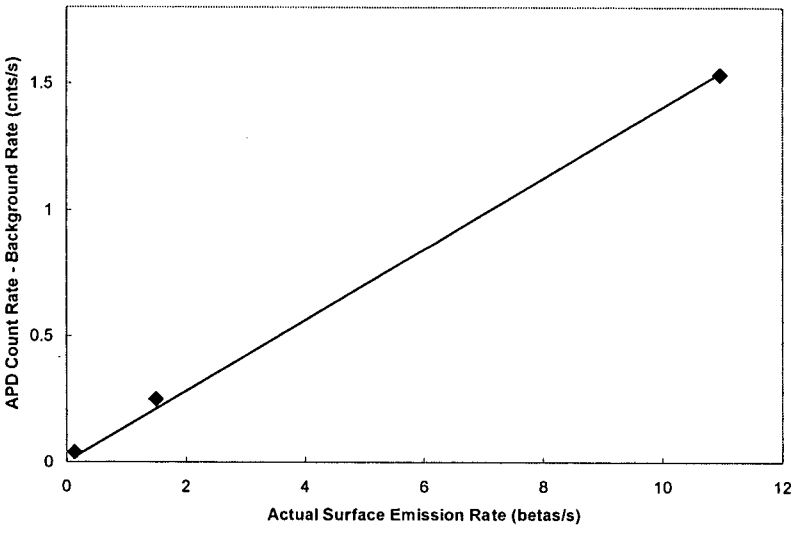

Figure 6 Comparison of (APD count rate - background rate) to actual beta emission rate

Also shown in Table 2 is a detection efficiency, i.e., the percent of betas emitted that are detected by the APD. The values shown, 14% to 29% are quite good when considering that significant signal was lost due to air attenuation and that the largest part of the tritium spectrum could not be counted due to excessive, low-energy background.

To evaluate the possibility that background measurements might depend on the non-tritiated material measured, background measurements were performed on plastic and paper as well as aluminum. These measurements are summarized with Table 3. All background measurements were similar. There are certainly no significant differences between aluminum and paper measurements. The background measured over plastic is larger than the other values, but this difference is small.

Table 3 Results of background measurements over various materials

Material Measured	Count Time (hours)	Integrated Count Rate, CN 300-100 (cnt/s)
Aluminum	44.2	0.041
Aluminum	3.3	0.044
Plastic	73.4	0.052
Paper	4.7	0.041

Discussion

Based on these measurements the APD system appears to be a very promising technique for determining tritium surface concentration. It is particularly important that it appears that the technique will be able to measure tritium surface concentration at the lowest of the free release limits used in the US (1000 dpm/100 cm^2). Using the prototype system and the method described here, a surface with this tritium concentration counted for two minutes would result in 10 counts. Five of these counts would be attributable to background and the remainder would be indicative of a tritium surface concentration at the lower free release level. Pending further validation, it may be concluded that this is sufficient to determine whether a surface is above or below the lower free release limit. This result would be obtainable in quite convenient period of time, especially compared to the time taken to perform scintillation counting preparation and measurement.

There are a number of points to keep in mind when considering use of an APD system for facility tritiated surface characterization. The traditional method of swipe/scintillation changes the surface through the act of swiping the surface, while the APD does not disturb the surface. Swipe/scintillation characterizes removable contamination, while the APD measures tritium at or very near the surface whether the tritium is removable or not. Besides direct measurement of a surface, an APD system can also be used to measure tritium on swipe. Given these facts, there are a number of scenarios that can be envisioned for an APD system. For instance, an APD could be used first to measure the surface. Then the surface could be swiped, and the APD could be used to quickly measure the swipe. In this fashion, both the fixed and removable tritium could be measured. One of the most attractive possibilities for an APD system is the possibility that this direct reading instrument could be placed at a radiological boundary so a quick determination could be made regarding whether or not items can be cleared out of the radiological area. Instruments have long been available to make such quick determinations for other radionuclides, but these instruments have not been available for tritium.

It should be noted that an APD system has the potential to significantly reduce the amount of waste associated with tritiated surface characterizations. Each swipe/scintillation sample required a swipe, a vial and cocktail. The APD system would eliminate all of these except possibly the swipe.

The APD system as presently envisioned can only directly read flat surfaces the size of the APD or larger and surfaces that are accessible. In cases where the APD cannot be used directly, it can still be used to indirectly characterize the surface by using swipes.

For appropriately shaped surfaces, an APD can be used to map surface tritium by rastering the detector across the surface. This method could be particularly useful for contamination/decontamination studies since the APD does not change the surface like a swipe would.

Further work is planned for APD development. The next steps will be testing of alternate APD configurations and testing APD's alongside swipe/scintillation in tritium facilities. There are a number of possibilities for increasing the sensitivity and usability of the APD.

Conclusions

A 77 mm^2 prototype APD system was tested with a view to directly characterizing surface tritium concentrations down to the US free release limits. It was found that:

- The APD can accurately characterize tritium surface concentration at the lowest of the US free release limits, i.e., 1000 dpm/100 cm^2. It appears that measurements at this level can be obtained in a mater of minutes.
- The APD was found to be very convenient to use. It was stable at room temperature, operated in normal room air, required no surface preparation and only required minimal effort to exclude ambient light from the APD surface.
- Excluding the low energy portion of the detection spectrum, the APD background was found to be very low. This low background was key to achieving the target sensitivity.
- Across three orders of magnitude of surface tritium concentration, the APD response was found to be essential linear and a calibration curve for the prototype APD was easily ascertained.
- As used in these tests, the only waste from APD operations was plastic placed between the APD and the measured surface.
- The APD was found to be stable with backgrounds and tritiated surface measurements staying the same from day to day.
- Various scenarios were identified for using APD's for tritiated surface characterization
- The efficiency (comparison of betas emitted to betas detected) of the APD system was 14% to 29%.

The authors found the APD system to be a very promising method for improving on current techniques used for tritiated surface characterization.

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